Construction and Building Materials 168 (2018) 995–1002

Contents lists available at ScienceDirect

Construction and Building Materials

journal homepage: www.elsevier.com/locate/conbuildmat

**Special Issue**
Use of NORM-containing products in construction

# Measurement of radioactivity in building materials – Problems encountered caused by possible disequilibrium in natural decay series

Boguslaw Michalik [a,*], Govert de With [b], Wouter Schroeyers [c]

[a] Główny Instytut Górnictwa (GIG), Plac Gwarków 1, 40-166 Katowice, Poland
[b] Nuclear Research and consultancy Group (NRG), Utrechtseweg 310, P.O. Box 9034, NL-6800 ES Arnhem, The Netherlands
[c] Hasselt University, CMK, NuTeC, Nuclear Technology – Faculty of Engineering Technology, Agoralaan building H, B-3590 Diepenbeek, Belgium

## HIGHLIGHTS

- Disequilibrium situations in uranium and thorium decay series are discussed.
- Assumption necessary to analyze gamma spectra of natural radionuclides are listed.
- Possible variations of activity index are showed.
- Related uncertainty in exposure assessment are underlined.

## ARTICLE INFO

*Article history:*
Received 7 July 2017
Received in revised form 6 February 2018
Accepted 8 February 2018
Available online 2 March 2018

*Keywords:*
Natural radioactivity
Gamma spectrometry
NORM
Index formula

## ABSTRACT

The determination of the activity concentration of naturally occurring radionuclides in construction materials is based on the principles of gamma-spectrometry. Gamma spectrometry is a comparative method and therefore includes many parameters that are specific to the test sample and measurement circumstances. Consequently, several of the testing conditions must be verified prior to testing and/or require correction to obtain accurate results. Besides problems encountered during the measurement, the interpretation of the results and calculation of the activity indices, needed for material classification, may lead to significant mistakes. Current regulation in the European Union requires to calculate an activity concentration index (index I) using the activity concentration of $^{226}$Ra, $^{232}$Th and $^{40}$K. Not all of these radionuclides are directly measurable by gamma spectrometry and, to determine the index, additional assumptions have to be made about secular equilibrium in uranium and thorium decay series. These assumptions are often not valid in case of NORM (Naturally Occurring Radioactive Materials) where long term lack of secular equilibrium in the uranium and/or thorium decay series is often observed. As a consequence, this may result in an underestimation or overestimation of the index. The article discusses specific disequilibrium situations in building materials. Sources for potential inaccurate determinations and misinterpretation are identified and practical mitigation options are proposed.


## 1. Introduction

Most construction materials are made from natural minerals. However currently many industrial residues are also used for this purpose. Specific residues can contain increased concentrations of naturally occurring radionuclides and for some of these residues, classified as NORM (Naturally Occurring Radioactive Materials), the related additional exposure to ionizing radiation is often not negligible. This problem was elucidated recently, to a certain level, by requirements set in Directive 2013/59/EURATOM, the so called Euratom Basic Safety Standards (EU BSS) [6]. This directive contains an indicative list of building materials that might be of concern from the radiation protection perspective and a general rule limiting additional exposure to ionizing radiation from building materials (less than 1 mSv per year expressed as effective dose). As it is not possible (or difficult, at least) to estimate the annual effective dose to members of the public caused by building material directly, the EU BSS offers a possibility to comply with the dose limit by limiting the radionuclides activity concentration in building materials and introduces a screening tool, the index I, to classify building materials from the radiation protection perspective.

* Corresponding author.
*E-mail address:* bmichalik@gig.eu (B. Michalik).

https://doi.org/10.1016/j.conbuildmat.2018.02.044
0950-0618/

This index, in fact the first time proposed in Radiation Protection 112 [5], is expressed as a weighed sum of activity concentration of chosen naturally occurring radionuclides:

$$I = \frac{C_{Ra-226}}{300} + \frac{C_{Th-232}}{200} + \frac{C_{K-40}}{3000} \leqslant 1 \quad (1)$$

where $C_{Ra\text{-}226}$, $C_{Th\text{-}232}$ and $C_{K\text{-}40}$ are the activity concentrations in Bq/kg of the corresponding radionuclides.

The weighting factors applied were obtained based on generic assumptions: (i) a dose criterion of 1 mSv/y – taken as an excess to the average background originating from the Earth's crust –, (ii) an annual indoor occupancy factor of 7000 h, and (iii) a conversion coefficient 0.7 Sv/Gy. The calculation of the weighting factors was based on a simplified room model i.e. room dimensions 4 m × 5 m × 2.8 m where surfaces (walls, floor and ceiling) were made of the same material (concrete of density = 2350 $kgm^{-3}$ and thickness of 0.2 m) without the presence of windows or doors. These calculations do not accurately reflect existing situations, instead the index *I* should be considered a conservative screening tool. This aspect is also underlined in the EU BSS by demanding a dose calculation that assumes specific parameters adapted to the intended use of the materials in the case of an index I larger than one.

Measurements of radionuclides are necessary to verify the compliance of building materials with the requirements of the European BSS. Determination of the activity concentration of natural radionuclides in construction materials is usually done by using gamma-spectrometry. This spectrometric technique is a comparative method and therefore includes many parameters that are specific to the test sample and measurement circumstances. Consequently, many of the testing conditions must be verified prior to testing and/or require correction to obtain accurate results. Moreover, not all radionuclides used for index I calculation are directly measurable by gamma spectrometry. That is why additional assumptions must be made about secular equilibrium in the uranium and thorium decay series. These assumptions are difficult in case of NORM where long-term lack of secular equilibrium in the uranium and/or thorium decay series is often observed and lack of the proper interpretation of the results may lead to significant mistakes. In spite of the fact that the system of differential equations describing sequential radioactive decay was already formulated by Rutherford [23] and relevant solution were proposed by Bateman [1] more than 100 years ago the possible influence of the lack of secular equilibrium on the final exposure is rarely considered when natural radionuclides are evaluated as a source of radiation risk.

In the current article $^{40}K$ is not discussed. The reason is that there is no problem to measure this radionuclide by gamma spectrometry based on single, efficient gamma line at 1462 keV or by classical chemical techniques, assuming its natural abundance of 0.0117% in natural potassium.

## 2. Disequilibrium in natural decay series

In raw materials, residues and building materials the members of the three natural decay chains, starting from $^{238}U$, $^{235}U$ and $^{232}Th$ can be present as well as $^{40}K$. As the ratio of $^{235}U$/ $^{238}U$ activities in natural uranium is very small (0.046 and this ratio can be changed only in the nuclear fuel preparation process, that usually does not generate residues or waste useful for construction purposes), in the *I* index calculation the contribution originating from $^{235}U$-decay series is assumed to be negligible. Moreover, other naturally occurring radionuclides ($^{3}H$, $^{14}C$ or $^{87}Rb$) are generally considered of no importance, considering their radiological impact, in this case. The presence of artificial radionuclides, that can be incorporated into building materials due to environmental contamination caused by nuclear or radioactive accidents (e.g. $^{137}Cs$), lays outside the scope of the I index used in the EU BSS.

Finally, from the perspective of the possible radiation risk caused by construction materials, two natural decay series (namely, the uranium and thorium series) are the most important (Figs. 1 and 2).

The radionuclides constituting these decay series usually occur all together, but the ratio of their activity concentrations depends on the origin and type of material of interest. In the natural state and when the influence of external factors can be excluded, all the radionuclides in a decay series are in secular equilibrium

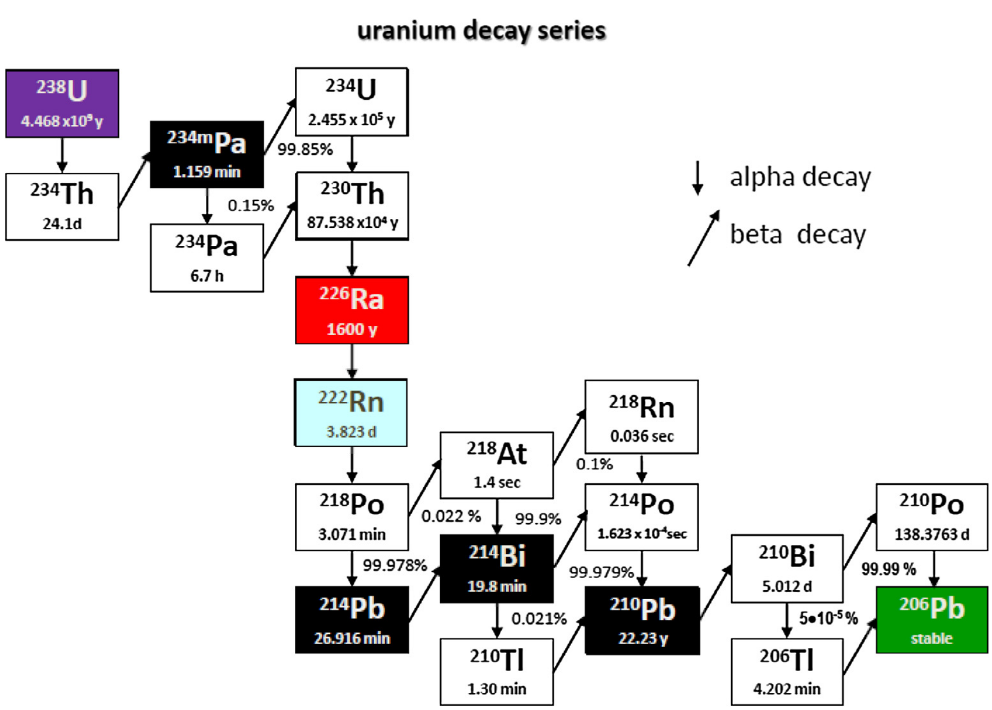


**Fig. 1.** Uranium decay series, in black boxes radionuclides emitting gamma radiation useful for gamma spectrometry data from Laboratoire National Henri Becquerel, [18].